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Progress in the First year

In the first year we focused on Multiphoton processes and their amplification in a nanocomposite medium to achieve upconversion lasing. This work was accomplished by collaboration with Professor Anderson Gomes ‘s group at Recife , Brazil . A significant research was accomplished and the resulting work was submitted to Optics Express , a high impact journal, for publication. We have received the reviewers’s comment. The two reviews who were asked to evaluate, have both given a positive review and asked for some revision. This work reports the operation and characterization of an upconversion random laser emitting at 560nm, when directly pumped by three photon excitation at the near IR wavelength of 1350nm in a colloidal dye solution in the weakly scattering regime. Using a special dye with a high three-photon cross-section and TiO_2 nanoparticles (250 nm diameter), optimized upconverted emission was obtained for particle densities of ~2 x $10^9/cm^3$. A strong dependence on the nanoparticle concentration and the pumping area was verified. The presence of spikes with linewidths ~ 0.4 nm in the emitted spectrum is the signature of coherent emission from this three-photon pumped random laser.

In our experiment (see Fig.1), a colloidal solution of the dye , 4-[N-(2-hydroxyethyl)-N-(methyl)amino phenyl]-49-(6-hydroxyhexyl sulphonyl) stilbene (APSS) dissolved in Dimethyl sulphoxide (DMSO) was mixed with commercially available TiO_2 nanoparticles (Dupont Inc, 250nm diameter) as the scattering media. The dye molarity was kept fixed at 0.06 mol/l. The colloidal RL sample was prepared by ultrasonically dispersing the nanoparticles (NP) with the laser dye. For the experiment, the suspension was placed within a quartz cuvette with 1cm x 1cm lateral dimensions. The pump source was a tunable optical parametric oscillator (OPO) pumped by a femtosecond (≈120 fs pulse duration) Ti:sapphire regenerative amplifier (Coherent Inc). The output of the OPO was tuned to the pump wavelength of 1350 nm, delivering pulses ≈100 fs, at variable repetition rate (up to 1 kHz) and a maximum pulse energy of 150 μJ. The pump beam was focused onto the cuvette using a 15 cm focal length lens, at an incident angle of 30^0, but the focus area on the sample was optimized for efficient RL emission. The RL emission was analyzed using a CCD coupled spectrometer (SpectraPro 300i, Princeton Instruments) with a spectral resolution of ≈0.1 nm. The scatterers provide the required feedback mechanism for RL emission, and the forward stimulated emission is suppressed.

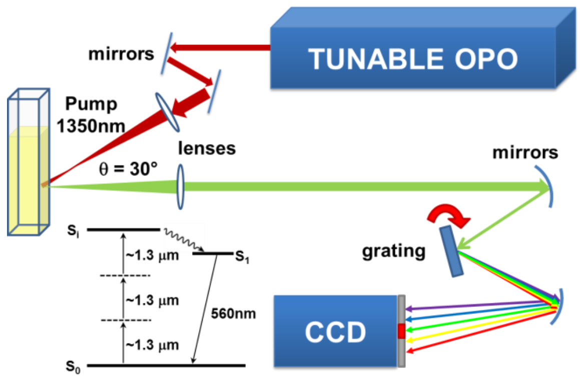

Figure 1 – Experimental diagram and energy level diagram for the three-photon excitation process in the APSS dye.

Figure 2 shows the spectral evolution at 1 kHz at various pump energies for a fixed excitation area of ~2.5 x 10^{-3} cm², and the spectral behavior for pump intensity well below and well above the RL threshold at 1 Hz. One can observe in both cases that the spectral width narrowing is quite striking. The typical linewidth reduction and nonlinear intensity dependence with the pump energy are shown in Figure 2. Results have been obtained for optimized NPs concentration and pumping area.

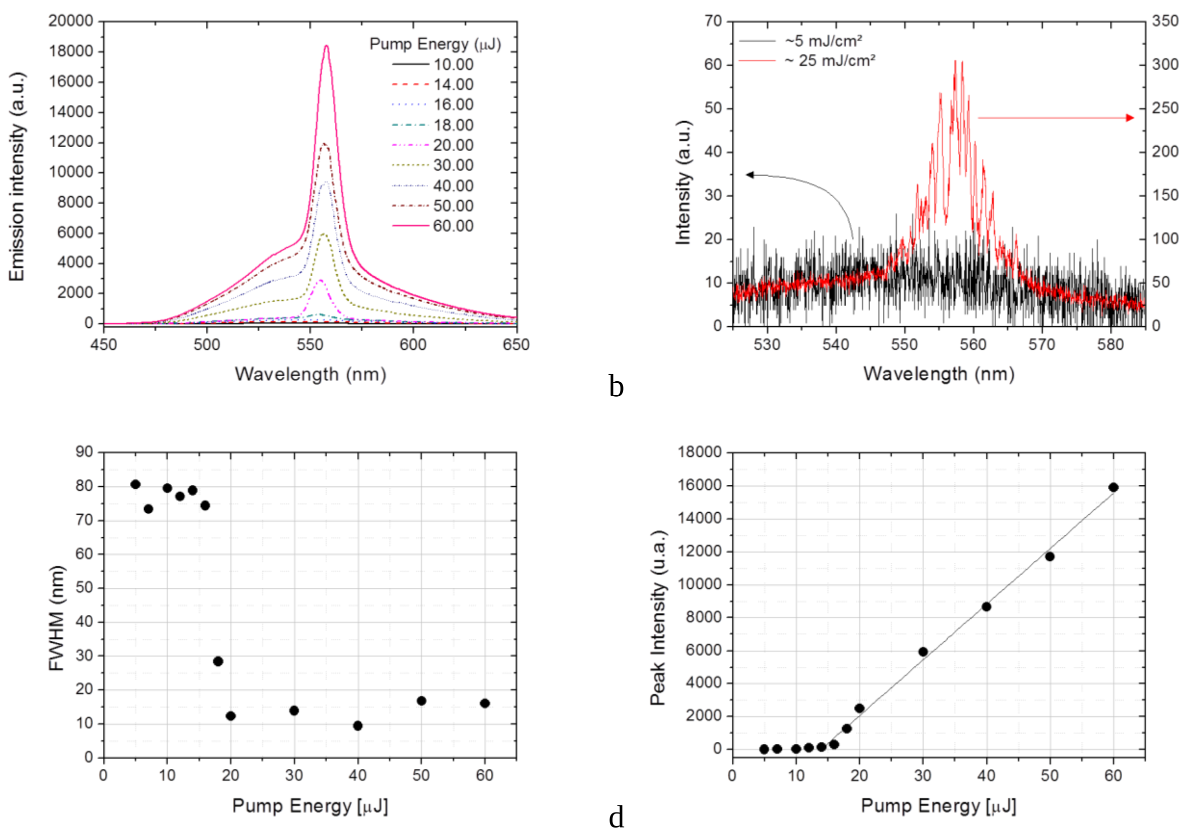

Figure 2 – (a) Spectral evolution of RL at 1 kHz for different incident pulse laser energy; (b) Single shot RL spectrum below and above threshold; (c) Linewidth reduction as a function of pump energy and (d) Emitted RL peak intensity as a function of pump energy.

As expected , there is a strong dependence of the RL intensity on the scatterer concentration and on the pumping area. The same qualitative behavior is demonstrated in our three-photon pumped system, as shown in Figure 3.

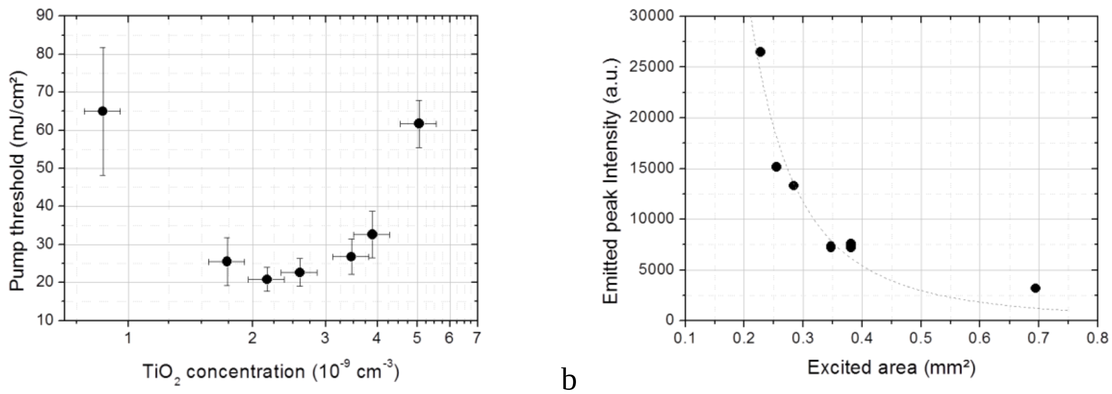

Fig 3 – (a) Pump threshold versus NP concentration and (b) emitted intensity as a function of pump area for the optimized concentration.

Results, obtained for pump excitation well above the threshold shown in Figure 4, clearly demonstrate the effect of averaging at a high (1 kHz in our case) repetition rate as compared to that at a low (<10 Hz) repetition rate. Although the spectrum at 1 kHz indicates the presence of spikes, such spikes are becoming very clearly resolved as the repetition rate is lowered. Fig. 4(b) shows a typical single-shot spectrum, which is very reproducible. The average linewidth of each spike is ~0.4nm.

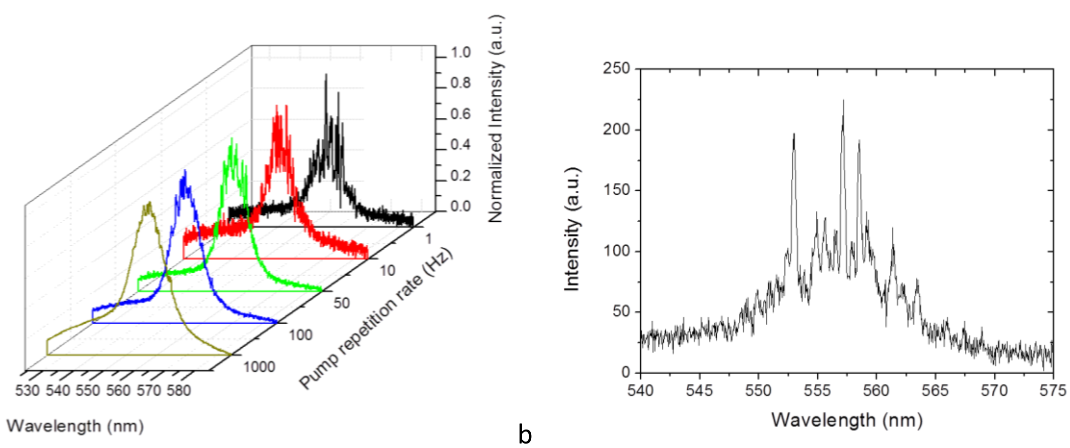

Fig. 4 – (a) Lasing spectrum obtained at different repetition rates; (b) Lasing spectrum at 1Hz. The pump excitation and pump area were 50µJ and 2.8 x 10^{-3} cm², respectively.

From the data in Figure 4(b), at the optimum concentration of $2x10^9/cm^3$, l_s is~ 1.75mm for our experiment (l_s ranged from 1.75cm to 0.6mm). Also, the so-called disorder parameter kl_t, where l_t is the transport mean free path, is very long and estimated to be ~ 43.000 at the optimum concentration. The result of Fig. 3 clearly demonstrates that the emission is dependent on scatterer concentration, while efficiency enhancement is a function of focusing.